

The Study on Segregation Purification for Nitrogen's Different forms through Constructed Wetlands

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Abstract-Nitrogen is one of the main pollutants in sewage. The traditional wastewater treatment technology, which lacks integrated collection systems, poses excessive investment costs on the control of nonpoint pollution. The constructed wetland is capable of removing nitrogen effectively; moreover, it is friendly to eco-system and requires low investment costs. So it is an important technology to reduce the nitrogen load, especially from the nonpoint source. In order to further study nitrogen removal mechanism and improve the removal rate of nitrogen from wastewater, combined floating bed and bio-film constructed wetlands (FBCW) simulation system was built to treat sanitary wastewater with filamentous palm, non woven and *Trifolium repens* L. TN, NH₃-N, NO₃-N, NO₂-N and pH were investigated for upper, middle and lower layer wetland system by intermission operation during 118 days under natural conditions. Results demonstrated that the system was into the stable phase of high removal rate after 48 days with hydraulic loading of 2m³/(m².d) and the hydraulic retention of 10 hours. Average removal rate of TN and NH₃-N were 64.4% and 60.6%, and the highest removal rate reached 79% and 77%, respectively. Segregation effect was clear. The purification ability of upper layer was higher than the middle. Middle layer was higher than the lower layer. TN and NH₃-N removal rate of upper layer were 1.96, 1.45 times and 1.36, 1.12 times higher than that of the middle and the lower layer, respectively. And the system has a good buffering capacity. pH variation of inflow water has a greater impact on the lower layer, while the upper pH values stabilized at 7.38-7.74.

Keywords-Constructed Wetlands; Bio-film; Floating Plant; Segregation Effect; Nitrogen

I. INTRODUCTION

With the rapid urbanization and industrialization, and highly accelerated economic development, increasingly serious environmental problems have ensued, especially in small and medium towns. Sewage has been discharged into water bodies directly without treatment and caused severe water pollution such as water eutrophication, which not only destroys the environment but also stunts its development. Large-scale centralized wastewater treatment systems have been regarded as a successful approach prevailing in

industrial countries in the last century in China^[1]. But these kinds of wastewater treatment technologies which lack integrated collection systems pose excessive investment costs on the control of non-point pollution. Complete replication of centralized wastewater treatment technology has proven to be rather limited and not entirely feasible.

Constructed wetlands (CWs) are artificial wastewater treatment systems consisting of shallow (usually less than 1 m deep) ponds or channels which have been planted with aquatic plants, relying upon natural microbial, biological, physical and chemical processes to treat wastewater^[2]. As a sustainable and low-cost and energy-efficient treatment method for wastewater^[3], many constructed wetlands have been commissioned to treat various types of wastewaters such as municipal and industrial wastewaters, urban and agricultural runoff^[4,5,6] and acid mine drainage. In addition, there is the advantage of multi-purpose re-use of the high quality effluent, self-remediation and self-adaptation to the surrounding conditions and environment^[7,8]. But the nitrogen removal effect of the constructed wetland needs to be improved especially for the sewage of lower carbon and higher nitrogen content. Many studies paid more attention to the nitrogen removal ability of constructed wetland in the sewage flowing direction not in the vertical direction. However, the nitrogen removal effect of the wetland was not only achieved by ammonia volatilization, media or plant absorption, but also the most vital factors of nitrification or denitrification by organisms. Previous studies showed that the nitrogen removed by denitrification reached 47~48% of the total nitrogen^[9]. Studies on segregation purification in the vertical direction could get better understanding of the nitrogen degradation mechanism inside the wetland system, supplying improved academic evidence for denitrification mechanism and the reference for engineering design. As there are so many papers on treatment of slightly polluted water and the eutrophication water focusing on the aquatic plant treatment craft^[10,11] at present, studies on combined treatment of plant floating and bio-film are hardly seen, as well as the internal segregation effect. For these reasons, the study reported the floating plant with bio-film constructed wetlands (FCW) for wastewater treatment focusing on the different layers of segregated purification effects of nitrogen.

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Constructed wetlands have been classified into two types by previous studies: Free water surface (FWS) and Vegetated submerged bed (VSB) systems (also known as subsurface flow wetlands), and single or integrated constructed wetland systems were built on this basis in many studies.

With the eutrophication of lakes, ponds and reservoirs, cultivation of plants on floating-bed to assimilate and remove the nutrients from water is an attractive phytoremediation approach^[12, 13, 14]. Floating beds have been applied widely to the treatment of eutrophic water. However, few reports and literatures on artificial floating bed technology were introduced to constructed wetlands. In particular, an understanding of Segregation Purification in floating plant with constructed wetlands (FCW) treatment systems is still lacking, as there are few researches on it.

This study reported on the performance of floating plant with bio-film constructed wetlands (FBCW) for wastewater treatment in 118 days and focuses on the different layer effects of nitrogen. The results provided successful cases and fundamental data for removal mechanism on Nitrogen and designing the reasonable size of constructed wetlands. This work differs from other investigations in the following aspects: (1) floating plant with bio-film constructed wetlands system was designed and constructed. (2) Removal mechanism on nitrogen segregation variation in domestic wastewater was analyzed in this system in lower and higher concentration, respectively.

II. MATERIALS AND METHODS

A. Construction of Simulation Wetland

The FBCW system (1000mm long, 150mm wide and 1000mm deep) was fabricated with 10mm thick PVC planks. There were three layers along the vertical direction in the system, which were plant floating layer, biological filter layer and precipitation layer. Non-woven fabric and palm silk filter, which were 150mm and 700mm high from the bottom, were used to divide the system into those three layers. Besides, the sampling pipes (A, B, C) in their respective layers were layout to do sampling.

B. Floating Plant

Trifolium repens L. was a perennial and xerophytes plant which can be easily got. It was chosen according to the preliminary study, and then transplanted and domesticated in the waste foam plastic. It could be used as the floating plant in wetland for its prosperous root system after domestication.

C. Bio-film Filler

Six biologic filling bars, which were woven with 1~2cm wide threads cut from non-woven fabric and palm fiber with 700mm of the overall length, were laid across the width of the system at intervals of 150mm.

D. Test Sewage

Test sewage was mixed wastewater from student canteen and toilets in lab building of Chengdu Textile College, and its water quality indexes were shown in Tab.1.

Table I EXPERIMENTAL WATER QUALITY (mg/L, EXCEPT pH)

Item	Concentration
TN	15-74.1
NH ₃ -N	10-50.6
NO ₃ -N	0.10-7.4
NO ₂ -N	0.03-0.43
pH	7.2-8.2

E. Model Experiments

The simulated experiment was carried out outdoor under natural conditions from April 5 to July 31 in 2011 in whole 118 days. The bio-film was formed when the surface of the fillers turned black naturally. The simulation system treated wastewater in an intermittent way. 300L wastewater in high cistern flowed into the bottom end of the constructed wetland and flowed out from the upper end continuously, with the hydraulic loading of 2m³/(m².d) and hydraulic retention time (HRT) of 10h. The whole cycle lasted 7 days: When the whole wastewater flowed into the constructed wetland, all the effluent was put back to the high cistern for repeated treatment after one day's suspension. After two day's suspension when last retreatment was over, new wastewater was treated in the system. Every sample was taken at the time when 2/3 wastewater had flowed across the wetland from the high cistern to ensure no wastewater left from the last cycle. Nutritional materials were added late in the experiment to verify the influence of pollution load on the removal ability of the simulation system. Ammonia nitrogen (NH₃-N), nitrate nitrogen (NO₃⁻-N), nitrite nitrogen (NO₂⁻-N), total nitrogen (TN), temperature and pH were measured according to Standard Methods (State Environmental Protection Administration of China, 2002).

III. RESULTS AND ANALYSIS

F. Water Temperature

The water temperature changes of inflow and outflow during the 118-day test period are shown in Figure 1.

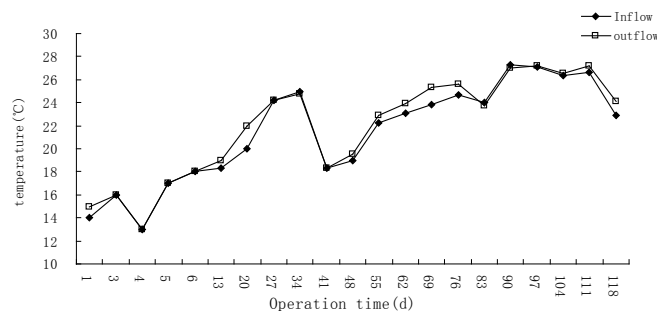


Fig. 1 Water temperature changes of inflow and outflow

Figure 1 shows the trend of temperature changes both in inflow and outflow water during 118 days. The water temperature gradually increased and the outflow were higher than the inflow. There might be reasons as follows: 1. The system of FBCW has some heat preservation and buffer

roles to temperature. 2. The strong biological activity was kept by the system from inside. 3. Organic matter and ammonia nitrogen, the oxidation of the pollutants also released a certain quantity of heat with aerobic-bacteria.

G. TN and $\text{NH}_3\text{-N}$

As bio-film has not formed yet in the early operation period, the simulation system was in an unsteady state where the effluent water quality changed greatly. Therefore, the nitrogen removal effect of the system in the vertical direction could be tested at the stable phase only. The variation curves of total nitrogen (TN) and $\text{NH}_3\text{-N}$ value were tested during the operation period within 118 days (Fig. 2 and Fig. 3).

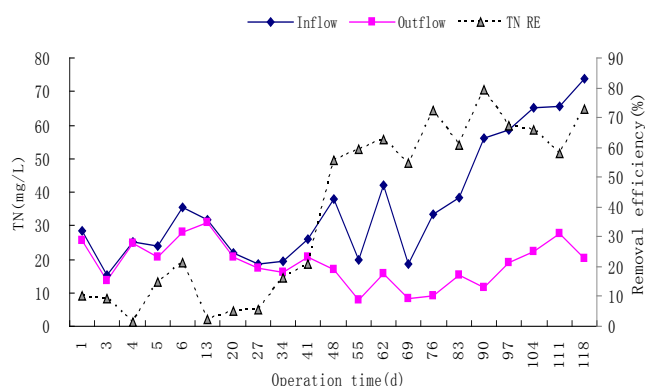


Fig. 2 TN changes of inflow and outflow

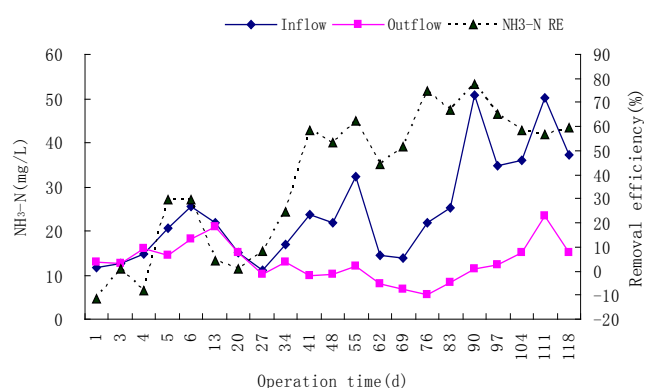


Fig. 3 $\text{NH}_3\text{-N}$ changes of inflow and outflow

Fig. 2 and Fig. 3 showed that the total nitrogen (TN) and $\text{NH}_3\text{-N}$ of inflow changed a lot during the 118 days, which was 15-74mg/L and 10-50mg/L respectively, but went to a sharp rise after 69 days' treatment. The outflow could be obviously divided into two phases. During the first phase (Day1-Day41), either the variation tendency of TN or $\text{NH}_3\text{-N}$ from inflow and outflow was consistent. The majority of the removal rate was below 20% only for several exceptions. Tests showed that the inflow and outflow had no significant differences ($P>0.05$). Besides, the concentration of $\text{NH}_3\text{-N}$ from outflow was even higher than the inflow in the first few days. During the second phase (Day 48-Day 118), no matter how inflow water quality changed, even with TN of 74mg/L, the TN and $\text{NH}_3\text{-N}$ of outflow could be remained relatively stable, which were 13-25mg/L and 11-21mg/L. The average removal rate of TN and $\text{NH}_3\text{-N}$ were 64.4% and 60.6%. With the nitrogen concentration of inflow increasing, removal rate of TN and $\text{NH}_3\text{-N}$ increased, while

the highest removal rate reached 79% and 77%. Tests showed that the water quality from inflow and outflow had significant differences ($p<0.01$). During the second phase, that the filters surface turned black demonstrated that bio-film was formed and the simulated system has already been in the stable phase. It took more time than any other researches for bio-film forming and stable phase entering^[15] because of natural bio-film forming without inoculation or air aeration. Data of 48-118 days were analyzed.

H. Segregation Effect on TN and $\text{NH}_3\text{-N}$

Nitrogen removal includes volatilization, ammonification, nitrification/denitrification, and plant uptake and matrix adsorption in wetland system. The inorganic nitrogen exploited by plant was a fraction of total nitrogen, while most of them were removed during the nitrification and denitrification^[16] through a synergy of biological, physical and chemical roles. This is a sophisticated biochemical process, referring to inter conversion among various nitrogen forms (Fig. 4), which was completed in the plant floating layer, biological filter layer and precipitation layer of FBCW system.

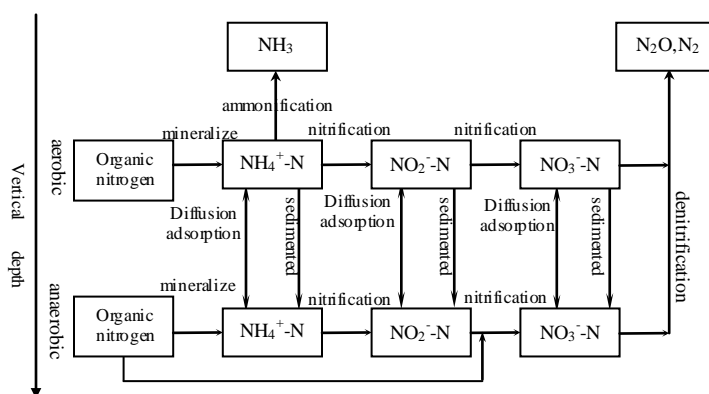


Fig. 4 Mutual conversion of nitrogen in constructed wetland

When the system was in the stable phase, the outflow water quality improved obviously and segregation phenomenon appeared at different depth. Fig. 5 and Fig. 6 showed TN and $\text{NH}_3\text{-N}$ concentrations of inflow and the segregation of the upper, middle and lower layers of the system during day 48 to day 118.

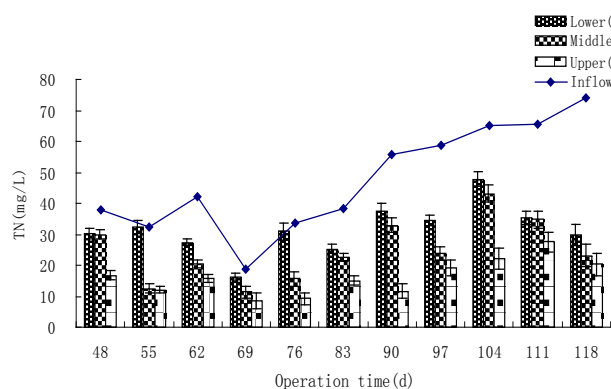


Fig. 5 TN segregation changes with difference depth

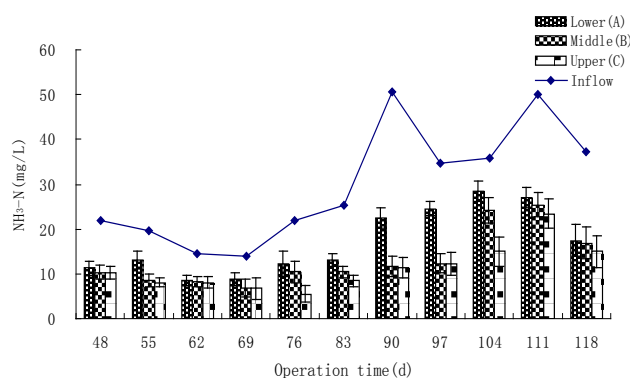


Fig. 6 $\text{NH}_3\text{-N}$ segregation changes with difference depth

As can be seen from Fig. 5 and Fig. 6, after 11 week's experiment, TN and $\text{NH}_3\text{-N}$ showed obvious changes of concentration gradient in different depth (A, B, C). As concentrations of TN and $\text{NH}_3\text{-N}$ were declining with the increasing of depth, the average TN concentrations from lower(A), middle(B), upper(C) were 31.5 mg/L, 24.5 mg/L, 16.2 mg/L during the 11 weeks while $\text{NH}_3\text{-N}$ concentrations were 16.9mg/L, 3.2mg/L, 11.3mg/L. When TN of inflow ranged from 30~40mg/L and $\text{NH}_3\text{-N}$ were at about 20mg/L, removing effect mainly happened in middle and upper areas. However, it happened in middle and upper areas while TN and $\text{NH}_3\text{-N}$ were up to 50 mg/L and 30 mg/L, respectively.

After domestic sewage flowed into the precipitation layer (lower layer) of the FBCW system, suspension would deposit because of filtration, sewage's gravity and also the nitrogen adsorption of the deposit. The average removal load of TN and $\text{NH}_3\text{-N}$ were 106.3mg/($\text{m}^2\cdot\text{d}$) and 84.5mg/($\text{m}^2\cdot\text{d}$) in this area. The nitrogen removal effect was unnoticeable while in lower concentration of inflow, but it turned to be obvious when concentration of inflow was increased. This could be related to the nitrification and denitrification in the later period of the area.

When sewage entered into biological filter layer, a large number of microbes' existence including facultative anaerobe, anaerobic microorganism and aerobe enhanced the nitrification and denitrification, as the average removal load of TN and $\text{NH}_3\text{-N}$ raising up to 152.9mg/($\text{m}^2\cdot\text{d}$) and 109.4mg/($\text{m}^2\cdot\text{d}$). The influence factors of removing TN was determined by that of $\text{NH}_3\text{-N}$ because the average $\text{NH}_3\text{-N}$ proportion of TN was 63% for domestic sewage. The ammonia nitrogen could not only be transformed to nitrate nitrogen and nitrite nitrogen in anaerobic environment^[16], but also could be ammonoxidation through *Nitrosomonas europaea* and *Nitrosomonas eutropha*^[17], and finally be removed as turning to N_2O or N_2 by denitrifying bacteria. In this area, it was the plenty micropores of non-woven fabric and palm silk in biological filter that supplied good environment for microbes to adhere and grow. Besides, palm silk is a cellulose fibre with high carbon content so that it could not only supply growth environment to microbes but also increase denitrification for carbon compensation during lack of carbon source and improve the nitrogen removal rate of the system.

Plants absorbed nitrogen as nutriment into its tissue, and also supplied proper place to nitrification and denitrification

in root zone in the nitrogen removal process. In the floating plant layer, *T.rpens* was able to fix nitrogen, and its domesticated flourishing root played a positive role in nitrogen removing in the system as the average removal load of TN and $\text{NH}_3\text{-N}$ were further improved to 208.5mg/($\text{m}^2\cdot\text{d}$) and 122.4mg/($\text{m}^2\cdot\text{d}$). It is worth noting that the removal effect of the floating plant area was decreased in later period of the process, as *T.rpens* was inhibited from growth for the increasing temperature and the rotted root because of the increasing sewage concentration as well as the addition of biological inhibitors.

From the analysis above, in our experiment system, no matter how changeable the nitrogen concentration was, the system had obvious segregation effect on removing total nitrogen and ammonia nitrogen. The effect of the upper layer was superior to the middle while the middle was superior to the lower. The average removal load of total nitrogen and ammonia nitrogen in upper layer were 1.96 and 1.45 times to the middle area while 1.36 and 1.12 times to the lower area. There into, the nitrification and denitrification of microbes in biological filter layer played an important role in nitrogen removing, and also the plant tissue absorption as well as the root zone supplying suitable environment for nitrification and denitrification.

I. Segregation effect on $\text{NO}_3\text{-N}$ and $\text{NO}_2\text{-N}$

As the domestic sewage was mixed wastewater from student canteen and toilets in lab building, it was found that ammonia nitrogen and organic nitrogen were the main nitrogen form after long-term monitoring and the concentration of nitrite nitrogen and nitrate nitrogen were low ranging from 0.04 ~ 0.1mg/L and 0.1 ~ 0.2mg/L. Therefore, when nitrite nitrogen and nitrate nitrogen were in low concentration, segregation was not obvious. On the contrary, when more potassium nitrate was added to increase the nitrate nitrogen concentration in the later period, the segregation effected obviously (Fig.7).

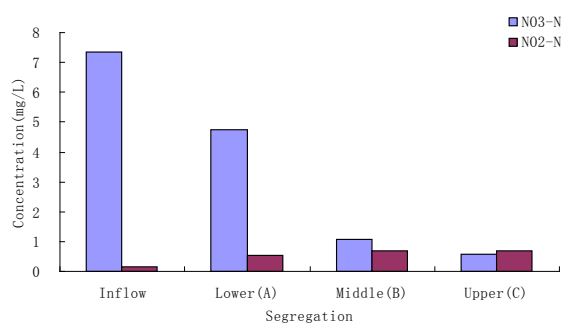


Fig. 7 NO_3NO_2^- segregation changes with different depths

From Fig.7 we could see, when the average concentration of nitrate nitrogen in inflow water was 7.4mg/L, the segregation purification for nitrogen of the system was achieved easily as the removal rate reached 92%. However, nitrite nitrogen concentration did not drop but increased considerably after the system treatment, and showed the segregation effects, which might be due to the further denitrification.

J. Segregation Effect on pH

Fig. 8 showed the variation of pH in the inflow and the upper, middle and lower layers in the FBCW system.

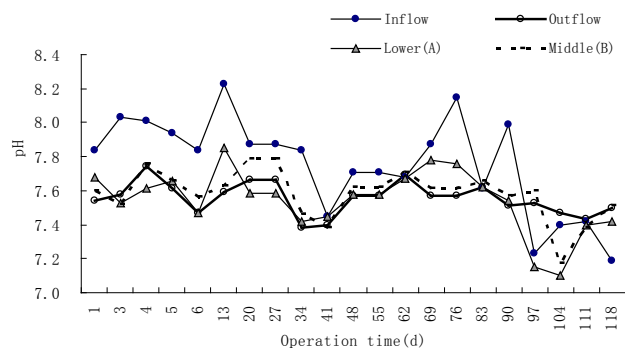


Fig. 8 Variations of pH in experimental cycle

As can be seen from Fig. 8, pH of inflow changed a lot (7.19-8.30), but when entered in the system, pH variation range was diminishing through the lower layer (7.10-7.90), the middle layer (7.17-7.79) and the upper layer (7.38-7.74) while the pH value of outflow remained stable. This demonstrated that the system has a good effect on buffering.

There might be some reasons as follows: the mineralization process could induce nitrate accumulation and hydrogen ion generation, and cellulose of palm silk decomposed produced lactic acid and acetic acid^[18], which caused pH value to decrease; meanwhile, reducing 1 mg nitrate nitrogen could produce 3.75 mg alkalinity^[19] through the denitrifying bacterium during denitrification, which could neutralize with the acidity produced by cellulose decomposition bacteria. But in the nitrification a great many of alkalinity was needed as oxidizing 1mg ammonia nitrogen could consume 8.64mg bicarbonate^[20], which could adjust pH value to some degree; at the same time, plant assimilated carbon dioxide from the outside atmosphere in the day time but assimilated oxygen and released carbon dioxide to the water in the night through their metabolism, so that this could lead to dissolved carbon dioxide increasing, which resulted in pH decreasing, especially in the upper layer.

IV. CONCLUSION

The FBCW system played a good buffering role as the pH value of outflow remained stable. The system remained stable after 48 days' operation, and the highest removal rate of total nitrogen and ammonia nitrogen went up to 79% and 77% with segregation effect. As for the bio-film in the middle layer, most nitrogen was decomposed through nitrification and denitrification. The purification effort of the middle layer was superior to the lower layer, and the upper layer was the best because of the synergistic effect of plant, nitrification and denitrification. The average removal load of total nitrogen and ammonia nitrogen at the upper layer were 1.96 and 1.45 times to the middle area while 1.36 and 1.12 times to the lower area. During the experiment, segregation purification of nitrate nitrogen and nitrite nitrogen were not obvious in the earlier period. The similar segregation effect

of nitrate nitrogen appeared as for the increasing nitrates nitrogen concentration of inflow in the later period, but the segregation effect of nitrite nitrogen were on the contrary.

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